

# Anomalous Behavior of the Upper Critical Field in Extreme Type-II Superconductors at Low Temperatures

Saša Dukan and Oskar Vafek\*

*Department of Physics and Astronomy, Goucher College, Baltimore, MD 21204*

## Abstract

We present a detailed numerical calculation of the upper critical field  $H_{c2}(T)$  for a bulk extreme type-II superconductor. Particular emphasis is placed on the high-field, low-temperature regime of the HT-phase diagram. In this regime it is necessary to go beyond the standard semi-classical theory and include the effects of Landau quantization of the electronic motion on the superconducting state. The presence of Landau level quantization induces an upward curvature in  $H_{c2}(T)$  at  $\sim 10\%$  of  $T_{c0}$  for those superconducting systems in which the slope of  $H_{c2}(T)$  at  $T_{c0}$  is  $\geq 0.2$  Tesla/Kelvin. We construct a simple analytical model that can account for this behavior based on the renormalization of the BCS coupling constant by the off-diagonal pairing of electrons on Landau levels.

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Corresponding author: Saša Dukan, Goucher College, 1021 Dulaney Valley Road, Baltimore, MD 21204

Phone: (410)337-6323, Fax: (410)337-6408, email: sdukan@goucher.edu

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\*Present address: Department of Physics and Astronomy, Johns Hopkins University, Baltimore, MD 21218

There has been considerable recent evidence of an anomalous "divergence" of the upper critical magnetic field  $H_{c2}(T)$  at low temperatures in a number of "low-temperature" high-temperature superconductors (HTS). Upward curvature and the anomalous divergence was observed in  $\text{Bi}_2\text{Sr}_2\text{CuO}_y$  [1] and  $\text{Tl}_2\text{Ba}_2\text{CuO}_6$  thin films [2];  $\text{K}_x\text{Ba}_{1-x}\text{BiO}_3$  single crystals [3,4];  $\text{Tl}_2\text{Mo}_6\text{Se}_6$  compound [5] as well in borocarbide intermetallic superconductors [6]. These observations are in contradiction with the standard semi-classical Werthamer-Helfand-Hohenberg (WHH) theory [7,8] which predicts the saturation of  $H_{c2}(T)$  at low temperatures. WHH theory yields a finite value of the upper critical field at zero temperature,  $H_{c2}^{WHH}(0)$ , and offers an elegant method for determining this field in clean superconductors as  $0.693T_{c0}(-dH_{c2}/dT)_{T=T_{c0}}$ , where  $(dH/dT)_{T=T_{c0}}$  is the slope of the upper critical field evaluated at the zero-field transition temperature  $T_{c0}$  (later in the text called the WHH-slope). For almost three decades, the WHH method has enabled experimentalists to determine  $H_{c2}(T)$  at low temperatures from the high temperature and low magnetic field data (around  $T_{c0}$ ) in conventional low-temperature superconductors. Recent advances in obtaining high magnetic fields in a laboratory environment [9] have revealed deviations of the experimental data from WHH theory at low temperatures.

WHH theory is based on the semiclassical phase-integral approximation originally due to Gor'kov [10] which depends on the following assumption: the bending of the semiclassical paths of the electrons by the magnetic field is negligible over the range of the single-particle Green's function at zero magnetic field. The latter is given by  $v_F/(2\pi k_B T)$ , while the radius of a semiclassical path equals  $l^2 k_F$ , where  $v_F$  and  $k_F$  are the Fermi velocity and wave vector, and  $l = \sqrt{\hbar c / e H}$  is the magnetic length. For a clean superconductor, this assumption reads  $l^2 k_F \gg v_F / (2\pi k_B T)$  or equivalently  $\hbar \omega_c \ll k_B T$ , where  $\omega_c = eH / m^* c$  is the cyclotron frequency. In dirty superconductors (with significant impurity concentration), it translates to  $\hbar \omega_c \ll \Gamma$  where  $\Gamma$  is the scattering rate due to disorder. From these conditions, it is clear that the semiclassical WHH theory ignores quantum Landau level (LL) effects, an assumption justified at low fields where the electrons occupy a huge number of closely spaced Landau levels (LL's). In this case, the temperature and/or impurity scattering broaden LL's

and reduce the significance of LL quantization. In the opposite limit of high magnetic fields and low temperatures, where  $k_B T \ll \hbar \omega_c$  and  $\Gamma \ll \hbar \omega_c$ , the quantization effects are of crucial importance. It was shown by Tešanović *et al.* [11] that the discreteness of the LL's leads to a breakdown of the semi-classical picture. When the LL level structure is fully accounted for in the BCS theory, one finds that, for a pure case or for a moderate level of impurities, superconductivity does not terminate above the semiclassical  $H_{c2}^{WHH}(T)$  line, but exhibits reentrant behavior where superconductivity is enhanced by the magnetic field  $H$  (where  $H \gg H_{c2}^{WHH}(T)$ ) [11,12]. A similar reentrant phase at high fields was recently proposed in organic quasi-one-dimensional superconductors [13]. As a consequence of the underlying LL structure  $H_{c2}(T)$ , or rather  $T_c(H)$ , develops oscillations near  $H_{c2}^{WHH}(0)$  signalling the passage of the LL through the chemical potential. Similar types of quantum oscillations, with the same origin, have been predicted in various other measurable quantities which are particularly pronounced in two-dimensional systems [12,14].

The LL quantization of electronic orbits *within the superconducting state* is now an experimental fact in numerous extreme type-II systems. The strongest evidence so far comes from recent observations of de Haas-van Alphen (dHvA) oscillations deep in the mixed state of various superconducting materials ranging from A-15's and boro-carbides to high- $T_c$  cuprates [15–17]. What all these materials (some of them also being samples with a “diverging” upper critical field) have in common is their extreme type-II character: the WHH slope in these systems is comparable to  $\sim 0.2$  Tesla/Kelvin and is often higher [1–6]. In such systems the cyclotron splitting of LL's near  $H_{c2}^{WHH}(0)$ ,  $\hbar \omega_{c2}(0)$ , where  $\omega_{c2}(0) \equiv e H_{c2}(0)/m^* c$ , is comparable to  $k_B T_{c0}$  and there is a large region in the H-T phase diagram in which the LL structure *within* the superconducting phase is well defined with  $\hbar \omega_c > \Delta(T, H)$ ,  $k_B T$  and  $\Gamma$  (here  $\Delta(T, H)$  is the BCS gap) [18]. The boundaries of this high field and low-temperature region in the  $H - T$  diagram,  $H^*$  and  $T^*$ , extend to fields as low as  $H^* \sim 0.5 H_{c2}(0)$  and temperatures as high as  $T^* \sim 0.3 T_{c0}$  [16]. In contrast, the size of this region in conventional type-II superconductors (like Nb) is negligible. Within the BCS theory, the scale of the cyclotron splitting between LL's near  $H_{c2}(0)$  in conventional systems is set by the condensa-

tion energy,  $\sim (k_B T_{c0})^2/E_F$ , and should be much smaller than either the thermal smearing,  $\sim k_B T$ , or the BCS gap  $\Delta(T, H)$ . Additional smearing due to the disorder  $\Gamma$  makes this high-field and low-temperature region in H-T diagram irrelevant.

In this paper we present a detailed numerical calculation of the upper critical field  $H_{c2}(T)$  for the extreme bulk type-II superconductor with particular emphasis on the high field and low temperature region in the H-T diagram. We assume a simple isotropic model based on the mean-field theory on LL's developed in our previous work [16]. We find LL structure inducing upward curvature in  $H_{c2}(T)$  at  $\sim 10\%$  of  $T_{c0}$  in these systems. In order to account for such behavior, we construct a scheme based on the renormalization of the BCS coupling constant by the off-diagonal pairing of the electrons on LL's. The model presented in this paper can be used to describe behavior of a cubic material such as  $K_x Ba_{1-x} BiO_3$  [3,4]. Furthermore, it can be extended to those anisotropic systems in which the hopping energy  $t^\perp$  is larger than a cyclotron gap at high fields, *i. e.*  $t^\perp > \hbar\omega_c$ . In such case, an isotropic model, with renormalized masses, correctly captures high-field behavior of a system. In the opposite limit of a highly anisotropic material or a quasi-two-dimensional system, mean-field theory is of a limited value due to strong fluctuations [11].

Within mean-field theory, the transition line  $T_c(H)$ , for any field  $H$ , is described as the solution of the self-consistent equation

$$\Delta(\mathbf{r}) = \frac{V}{\beta} \sum_{\omega} F(\mathbf{r}, \mathbf{r}; \omega) \quad (1)$$

when the amplitude of the superconducting order parameter  $\Delta(\mathbf{r})$  goes to zero.  $V$  is the usual BCS interaction strength and  $\beta = 1/k_B T$ .  $F(\mathbf{r}, \mathbf{r}; \omega)$  is the anomalous Green's function with  $\omega = 2\pi k_B T(m + 1/2)$  being the electron Matsubara frequency (the Matsubara index  $m$  is dropped in order not to be confused with the LL index later in the text). This anomalous Green's function is constructed from the wavefunction of the individual electrons in a magnetic field (for review see reference [19]). For a clean three-dimensional (3D) superconductor in high magnetic field equation (1) reduces to the form

$$1 = \frac{V}{2\pi l^2 \beta} \sum_{n,n'=0}^{\infty} \frac{(n+n')!}{n!n'!2^{n+n'}} \int \frac{dk_z}{8\pi} \sum_{\omega} \frac{1}{i\omega - \varepsilon_{n\pm}(k_z)} \times \frac{1}{-i\omega - \varepsilon_{n'\mp}(k_z)} \quad (2)$$

where  $n$  and  $n'$  are indices of the Landau levels participating in the superconducting pairing and  $k_z$  is the momentum along the field direction. The electronic energies in the magnetic field

$$\varepsilon_{n\pm}(k_z) = \frac{\hbar^2 \vec{k}_z^2}{2m^*} + \hbar\omega_c(n + \frac{1}{2}) \mp g \frac{\hbar e H}{4m} - \mu = \frac{\hbar^2}{2m^*}(k_z^2 - k_{Fn\pm}^2) \quad (3)$$

are measured from the chemical potential  $\mu$ . In the presence of Zeeman splitting ( $g \neq 0$ ), for each value of  $n$  and for each index “plus” and “minus” there is a corresponding Fermi momentum  $k_{Fn\pm}$  determined by the conditions (for  $T/\mu \ll 1$ )

$$k_{Fn\pm} = \sqrt{\frac{2m^*}{\hbar^2}(\mu - \hbar\omega_c(n + 1/2) \pm g \frac{\hbar e H}{4m})} \quad (4)$$

$$n_e = \frac{1}{2\pi^2 l^2} \sum_{n\pm} k_{Fn\pm} \quad (5)$$

where  $n_e$  is the electronic density and the sum is over all real  $k_{Fn\pm}$ . In deriving self-consistent equation 2 we assume that the order parameter  $\Delta(\mathbf{r})$  has a standard Abrikosov form [20]. In the LL representation Abrikosov order parameter belongs to the lowest Landau level (LLL) for the Cooper charge  $e^* = 2e$  having the lowest kinetic energy of the center-of-mass motion of Cooper pairs and therefore the highest transition temperature. Note though, that in two dimensions (2D) special circumstances might arise where the contributions to the order parameter from the higher LL's become competitive leading to the higher transition temperature [12]. However, in 3D, higher LL contributions lead to the significantly lower transition temperature [11,21].

In the framework of the BCS theory, the density of states (DOS) of the electrons in a magnetic field exhibits a divergence whenever the bottom of a LL crosses the chemical potential. As a consequence, strong oscillations in  $T_c(H)$  develop at high fields and low temperature in a clean superconductor [11]. These oscillations, in principle, could be observed in extremely clean samples which is rarely the case in an experimental setup. Therefore, we consider a more realistic system for which the presence of the impurities and imperfection

(or disorder in general) is taken into account. We assume that the disorder in the sample leads to isotropic broadening of the LL's, the size of which is measured by  $\Gamma = \hbar/2\tau$ , where  $1/2\tau$  is the scattering rate due to the disorder. As long as  $\Gamma/\hbar\omega_c \ll 1$  the discreteness of the LL structure is preserved but the quantum oscillations are greatly reduced. In this way we are left only with the task of numerically examining the overall rising trend of  $T_c(H)$  at high fields. The broadening of the LL can be most easily included into the self-consistent equation (2) by a substitution  $i\omega \rightarrow i\omega - \Gamma$  in the Matsubara frequencies.

After integration over the momenta  $k_z$  and summation over the Matsubara frequencies (with the Debye frequency  $\Omega$  as a UV cut-off), the equation (2) can be put in the form

$$\frac{1}{\lambda} = \sum_{n,n'=0}^{\infty} \frac{(n+n')!}{n!n'!2^{n+n'}} \frac{N_{1CM}(0)}{2\pi l^2 N_{3D}(0)} \frac{1}{4} [\Psi(Z_{1\pm}) + \Psi(Z_{1\pm}^*) - \Psi(Z_{2\pm}) - \Psi(Z_{2\pm}^*)]$$

$$Z_{1\pm} = \frac{\Omega}{2\pi T} + i \frac{\hbar\omega_c(n - n' \pm g/2)}{4\pi T}$$

$$Z_{2\pm} = \frac{1}{2} + i \frac{\hbar\omega_c(n - n' \pm g/2)}{4\pi T} \quad (6)$$

where for each value of the index  $n$  there are two terms, one for the “+” and one for the “-” sign.  $\lambda = VN_{3D}(0)$  is the BCS coupling constant with  $N_{3D}(0)$  being the single-spin electronic DOS in zero field calculated at the Fermi energy  $E_F$ .  $\Psi(z)$  is the Digamma function of the complex variable  $z$ , and  $z^*$  denotes complex conjugation.  $N_{1CM}(0)/2\pi l^2$  is the DOS of the center-of-mass of the Cooper charge at the chemical potential and it compares to  $N_{3D}(0)$  as

$$\frac{N_{1CM}(0)}{2\pi l^2 N_{3D}(0)} = \frac{\Gamma}{2^{5/2} \sqrt{E_F \hbar\omega_c} (C^2 + (\frac{\Gamma}{\hbar\omega_c})^2)^{1/2} (\sqrt{C^2 + (\frac{\Gamma}{\hbar\omega_c})^2} - C)^{1/2}} \quad (7)$$

where

$$C = \frac{\mu}{\hbar\omega_c} - \frac{n + n' + 1}{2} . \quad (8)$$

Our main goal is to numerically solve the self-consistent equation (6) for the transition temperature  $T_c(H)$  taking realistic values for the coupling constant  $\lambda$  as well as moderate values for the disorder parameter  $\Gamma/E_F$ . In particular, we want to incorporate in our calculation the crucial observation that the WHH-slope  $(-dH_{c2}/dT)_{T=T_{c0}}$  reported in the experiments

[1–6] is always much larger than  $\approx 0.2$  Tesla/K. Therefore, we chose a coupling constant  $\lambda$  so that the WHH-slope when expressed in the dimensionless units  $[-d\omega_c/d(k_B T)]_{T=T_{c0}}$  is larger than 0.27 [18].

Figure 1 shows the numerical solution of equation (6), *i.e.* the plot of the upper critical field  $H_{c2}(T)$  rescaled by  $H_{c2}^{WHH}(0)$  *vs.*  $T_c/T_{c0}$  for the model system with coupling constant  $\lambda = 0.35$  (full line) and  $\lambda = 0.4$  (dotted line), the disorder parameter  $\Gamma/E_F = 0.025$  and zero Zeeman splitting ( $g = 0$ ). For the model with  $\lambda = 0.35$  the WHH-slope in dimensionless units is  $[-d\omega_c/d(k_B T)]_{T=T_{c0}} = 0.8$  which corresponds to  $(-dH_{c2}/dT)_{T=T_{c0}} = 0.58$  Tesla/K. When  $\lambda = 0.4$ , the WHH slope in dimensionless units is 1.108, or 0.813 Tesla/K in conventional units. The upper critical field starts to deviate from the one predicted by the WHH theory at  $T_c/T_{c0} \approx 0.1$  exhibiting an anomalous divergence at low temperature as predicted by Tešanović *et al.* [11] and recently seen in experiments [1–6]. While disorder completely washes away the quantum oscillations of the upper critical field at low temperatures for  $\lambda = 0.35$  case, they are still visible for the model with the  $\lambda = 0.4$ .

In Figure 2 we plot the ratio  $\hbar\omega_c/E_F$  (which is proportional to  $H_{c2}$ , the proportionality constant being set by the material properties of the particular superconductor) *vs.*  $T_c/T_{c0}$  for the model system with  $\lambda = 0.35$  in order to illustrate the effect of Zeeman splitting ( $g \neq 0$ ) on the upper critical field. The overall tendency of the Zeeman splitting is to suppress  $T_c$  in the region of the H-T diagram where  $k_B T \leq \hbar\omega_c$ , while the transition line is not affected by Pauli pair breaking in the high temperature-low field portion of the diagram. An interesting situation arises when effective  $g$  factors are very close to  $g = 2m/m^*$ ,  $g = 4m/m^*$  or any other even integer (we take the effective cyclotron mass  $m^*$  of the order of the electron mass  $m$  further in the text). Unlike the odd  $g$  factor case ( $g = 1$  is an example shown in Figure 2), the anomalous divergence of the upper critical field at low temperature is not destroyed by Zeeman splitting for even integer  $g$  factors. The temperature at which  $H_{c2}(T)$  starts to deviate from the WHH line is reduced by only few percent from the  $g = 0$  case. This situation can be understood as follows: When  $g = 2$  or  $g = 4$  the Zeeman splitting is equal to the cyclotron splitting making the  $n$ th spin-up LL degenerate with the  $(n + 1)$ th (for

$g = 2$ ) or  $(n + 2)$ th (for  $g = 4$ ) spin-down level. In these cases the off-diagonal terms in equation (6), describing LL's separated by the cyclotron gaps, become effectively diagonal (*i.e.* degenerate) leading to the anomalous divergence in  $H_{c2}(T)$  (see the discussion below). When  $g$  is an odd integer or a fraction, the LL spin degeneracy is completely lifted resulting in the suppressed  $H_{c2}(T)$  at low temperatures with a downward curvature.

The upward curvature of the upper critical field  $H_{c2}(T)$  at temperatures  $T < 0.1T_{c0}$  is the consequence of the competing tendencies of diagonal,  $n = n'$ , and off-diagonal,  $n \neq n'$ , terms in the self-consistent equation (6). The diagonal terms correspond to the Cooper pairs formed by electrons in the same LL while the off-diagonal terms represent the electronic pairing of the LL's separated by  $\hbar\omega_c$  or more. Only the diagonal terms possess Cooper singularity and therefore lead to the increasing trend of the transition temperature as a function of field [11,12,14]. In clean systems, they also produce strong quantum oscillations in  $T_c(H)$  for fields  $H > H_{c2}^{WHH}(0)$ . Ultimately, the diagonal terms in (6) lead to the reentrant superconductivity for magnetic fields  $H \gg H_{c2}^{WHH}(0)$ . This Quantum Limit regime, reached when all electrons in the system occupy a single, lowest Landau level, was predicted and investigated by Tešanović *et al.* [11] but is not of interest in our calculation since it can be realized only in extremely high fields. The off-diagonal terms in (6) start to dominate at lower fields  $H \approx H_{c2}^{WHH}(0)$  creating a counter-effect to the rapidly decreasing diagonal terms. These *non-singular* terms lead to the smooth crossover from the diverging high-field transition line to the low-field WHH line. This crossover results necessarily in the upward curvature of the upper critical field  $H_{c2}(T)$  at low temperatures. Our goal in this paper is to closely examine this crossover behavior of the off-diagonal terms and to account for the resulting upward curvature in a simple analytic model. First, we notice that the effective role of the off-diagonal terms in the self-consistent equation (6) is to *renormalize* the BCS coupling constant  $\lambda$  into a new, field and temperature dependent, constant  $\tilde{\lambda}(H, T)$  through the substitution  $1/\lambda \rightarrow 1/\lambda - 1/\lambda'(H, T) = 1/\tilde{\lambda}(H, T)$  where  $1/\lambda'(H, T)$  accounts for the off-diagonal,  $n \neq n'$ , terms. The size of the off-diagonal terms in (6) grows as the magnetic field is lowered, leading to the effective increase in the renormalized BCS coupling constant

$\tilde{\lambda}(H, T)$  rendering a non-zero transition temperature at lower fields.

With this approach the solution of the self-consistent equation can be written in analytical form as

$$T_c(H) = 1.134\Omega \exp \left[ -\frac{2\pi l^2}{\tilde{\lambda}(H, T_c)} \left[ \sum_{n=0}^{n_c} \frac{N_{1n}(0)(2n)!}{N_{3D}(0)2^{2n}(n!)^2} \right]^{-1} \right] \quad (9)$$

where  $N_{1n}(0)$  is the 1D density of states given by (7) and (8), where  $n = n'$ . When  $2\pi k_B T < \hbar\omega_c$  the contribution of the off-diagonal terms  $1/\lambda'(H, T_c)$  to the renormalized coupling constant can be obtained by systematic expansion in  $2\pi k_B T_c/\hbar\omega_c$ . Assuming that the number of the occupied LL's  $n_c = E_F/\hbar\omega_c$  is much larger than one, *i.e.* that the system in question is far away from the Quantum Limit regime (in a typical experimental setup this is indeed the case, since usually  $n_c > 30$ ), and ignoring the quantum oscillations (they are damped by disorder), we perform the expansion of the off-diagonal terms up to  $(2\pi k_B T_c/\hbar\omega_c)^2$ . Then, it is possible to rewrite the equation (9) in very simple analytic form as

$$T_c(H) = 1.134\Omega \exp \left[ -\frac{4\sqrt{n_c}}{\sqrt{\pi}} \left( \frac{1}{\lambda} - \frac{1}{\lambda'(H, T_c)} \right) \right] \quad (10)$$

where

$$\begin{aligned} \frac{1}{\lambda'(H, T_c)} = & \exp \left( -\frac{2\sqrt{\pi}}{n_c} \right) + \exp \left( -\frac{1}{2\sqrt{\pi n_c}} \right) \ln \sqrt{n_c} + \frac{1}{2n_c} \left( \frac{2\pi k_B T_c}{\hbar\omega_c} \right) \\ & + \left( \frac{2}{3\pi\sqrt{n_c}} + \frac{1}{3\pi\sqrt{n_c^3}} - \frac{\pi^2}{6n_c^2} \ln(n_c) \right) \left( \frac{2\pi k_B T_c}{\hbar\omega_c} \right)^2. \end{aligned} \quad (11)$$

Equation (10) can be solved by iteration or can be used as a formula to fit the experimental data. Figure 3 shows  $T_c(H)$  computed from (9) with  $1/\lambda'(H, T_c)$  in (11) expanded to leading order (dotted line) and to second order (dashed line) in  $2\pi k_B T_c/\hbar\omega_c$  compared to the exact numerical solution of self-consistent equation (6) for  $\lambda = 0.35$ . The agreement is excellent in the region where  $2\pi k_B T < \hbar\omega_c$ , where the expansion (11) is valid. The simple analytic form (10) accounts very well for the diverging upper critical field  $H_{c2}(T)$  and its anomalous upward curvature. When  $2\pi k_B T = \hbar\omega_c$  this expansion breaks down as indicated in Figure

3 by the straight line. At low fields and high temperature, where  $2\pi k_B T > \hbar\omega_c$ , there is a large deviation of  $T_c(H)$  from the exact numerical solution of (6), signalling the breakdown of the expansion.

To conclude we have presented a detailed numerical calculation of the upper critical field  $H_{c2}(T)$  for a three dimensional extreme type-II superconductor characterized by a large WHH slope. We find that the Landau level quantization induces an upward curvature in  $H_{c2}(T)$  at temperatures  $\sim 0.1T_{c0}$ . We account for this behavior through renormalization of the BCS coupling constant  $\lambda$  by the off-diagonal pairing of the electrons in the Landau levels. Our work, based on the simple BCS model, reproduces qualitatively observations of the anomalous behavior of the upper critical field in the experiments [3,4], but it cannot account quantitatively for the large deviations in  $H_{c2}(T)$  from the WHH line for temperatures higher than predicted by our theory. We believe that extension of this work to a more realistic strong-coupling model within the Landau level framework will improve agreement with the experimental results.

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## FIGURES

FIG. 1. Plot of the upper critical field  $H_{c2}$  computed from (6) vs.  $T_c/T_{c0}$  for a 3D superconductor with the BCS coupling constant  $\lambda = 0.35$  and  $\lambda = 0.40$  and with no Zeeman splitting.  $H_{c2}$  is rescaled by  $H_{c2}^{WHH}(0) = 0.693T_{c0}(-dH_{c2}/dT)_{T=T_{c0}}$ .

FIG. 2. Upper critical field  $H_{c2} \sim \hbar\omega_c/E_F$  for a 3D superconductor with non-zero Zeeman splitting.

FIG. 3. Comparison of the exact numerical solution of (6) (full line) and the solution of (9) obtained by the expansion of the off-diagonal terms in (6) up to the leading (dotted line) and the second order (dashed line) in  $2\pi k_B T_c/\hbar\omega_c$ . To the left of the straight line  $2\pi k_B T_c = \hbar\omega_c$  this expansion is valid.





